

a coating, or as discrete particles embedded in electrodes. Electrodes carry the current generated.

[0070] In all fuel cell types, the natural properties of electrolyte materials determine the speed of travel of the charged particles. In other words, nature determines, to a large extent, the speed of charged particle transport across the electrolyte. In this invention, charged particle transport characteristics of electrolyte material is enhanced, first by shortening the solid state transportation path by providing pores within electrolyte; and second by increasing the defect concentration of solid oxide membranes. In doing so, the manufacturing of solid oxide membranes is simplified as well, by first forming a metal membrane, and next oxidizing the metal to form an oxide electrolyte membrane.

[0071] The electrolyte material can be used more efficiently if it were a continuous structure with oxygen and hydrogen carrying gas flow channels traversing it. And, if the electrolyte were also to act as the structural member of the fuel cell providing support for all other components of the fuel cell, i.e., electrodes and catalysts, then, it would be relatively easy to reduce the electrolyte thickness between oxygen and hydrogen carrying channels, adjust the size of channels, as well as the distance between them to increase the reaction surface area. It is understood that the overall efficiency of a fuel cell depends on the reaction surface area and the thickness of the electrolyte the charged particles must travel, provided that all other material related factors remain the same. Thus, fuel cells based on the three-dimensional electrolyte concept of this invention can be several orders of magnitude more efficient than the conventional two-dimensional fuel cells.

[0072] Fuel cells have advantages over conventional combustion-based technologies used in power plants and cars. They produce very little greenhouse gases and no smog and illness causing air pollutants. If pure hydrogen is used as a fuel, fuel cells emit only heat and water as byproducts.

[0073] This invention also provides two improved membranes for fuel cells, and a method of manufacturing. One is a solid electrolyte that is manufactured initially from a metal, and later reacted to form a solid (oxide) electrolyte, at least partially, during manufacturing of the fuel cells. Here the solid electrolyte maybe an oxide, sulfide, bromide, chloride or any other substance that allows charged particle conduction, but little or no electronic conduction.

[0074] Although, the electrolytes of this invention can be used in conventional, two-dimensional, fuel cells, as those used in conventional fuel cells, the preferred fuel cell design is the novel three-dimensional fuel cell design, which will be described next. Three-dimensional fuel cells of this invention consist of rows of closely spaced channels within the electrolyte matrix, each alternately carrying fuel gas and oxygen containing gas, separated by very thin electrolyte. The electrolyte is a continuous medium produced in dimensions sufficient to create desired electrical energy; said medium also acts as load carrying construction material. Electrodes are located within the gas flow channels, and thus maybe referred as electrode/channel in this specification. Electrode material may contain a catalyst on or near its surface, either as a coating, or as discrete particles embedded in electrodes. Electrodes carry the current generated.

[0075] Charged particle transport medium in this invention is referred to as electrolyte and or membrane, often interchangeably.

[0076] FIG. 1 is an external view of a hydrogen fuel cell power system 10 according to this invention, where gas inlet pipes 14 and 15, and gaseous exhaust pipes 16 (hidden in the drawing of FIG. 1), and 17 are shown. At each corner of the power system, there are two primary gas inlet channels 18 and 13, and two primary exhaust channels 11 and 12. An electrical receptor 13 is mounted at one corner. At the receptor 19, electrons generated through reactions within the cells are available to an outside circuit. The shape of the hydrogen fuel cell power system 10, whether there is an outside cover plate or not, size and locations of inlet-outlet pipes, their shapes, shape and size of primary inlet and outlet (exhaust) channels are depicted as examples only, and could differ without any significant effect on the description of the device offered here.

[0077] The plane AA' shown in FIG. 1, intersects many gas flow channels, including light colored channels 22, which carry oxygen-containing gas, and darkened channels 23, 24, 25, and 26, which carry hydrogen-containing gas as shown in FIG. 2. In this figure, only a small number of possible channels are shown. Porous electrodes within the channels are not shown for simplicity of explanation. Also, the channels are shown in black and white in order to differentiate between channels carrying gas containing oxygen 22 and channels 23, 24, 25, 26 carrying hydrogen-containing gas.

[0078] Referring to FIG. 2, each oxygen-carrying channel 22 communicates with its nearest neighbor hydrogen carrying channels 23, 24, 25, and 26. Similarly, each hydrogen-carrying channel communicates with four of its nearest neighbor oxygen-carrying channels. Thus, each gas flow channel sends or receives charged particles across the electrolyte 29. Accordingly, the number of cells in a hydrogen fuel cell power system, according to this invention, is ideally equal to the number of gas flow channels divided by two. Since, each gas flow channel communicates with four of its nearest neighbor channels, reaction surface of each channel is its entire surface. To form a fuel cell, there needs to be no more than two channels with electrodes, one carrying oxygen and the other hydrogen containing gas. Obviously, more channels there are, the higher are the energy level developed by the fuel cell.

[0079] Gas flow channels shown in FIG. 2 extend some distance within electrolyte 29 as desired, or as gas pressure requirements dictate. In FIG. 3, a three dimensional view of the gas flow channels are shown. Again for the sake of providing a simple description, channels carrying hydrogen and oxygen are shaded dark and white respectively, and the electrodes are not shown. Hydrogen carrying tertiary channel 31, and other similar tertiary channels in the same row receive their gas from a larger secondary gas flow channel 33. Oxygen carrying tertiary channel 35, and other similar tertiary channels in the same row receive their oxygen-carrying gas from a larger secondary gas flow channel 37. Referring to FIG. 4, the secondary gas flow channels 37 join a larger gas flow channel 18, which is the primary gas flow channel and is large enough to feed all the secondary gas flow channels. There is a corresponding primary hydrogen-containing gas flow channel 13 feeding all secondary hydrogen-containing gas flow channels. This is shown in FIG. 1.

[0080] Referring to FIG. 3 again, hydrogen-carrying tertiary channel 31, and other similar tertiary channels in the same row exhaust their gas from a larger secondary gas flow channel 39 at the other end. Oxygen-carrying tertiary chan-

polymers C—S bonds in the main chain, examples being polysulfide ethers, polyphenylene sulfide, polyether sulfone, polysulfone, polyether ether sulfone, polyaryl ether sulfone, polyphenylene sulfone, polyphenylene sulfide sulfone and poly(phenyl sulfide-1,4-phenylene);

polymers C—N bonds in the main chain, examples being polyimines, polyisocyanides, polyetherimine, polyetherimides, poly(trifluoromethyl)bis(phthalimide)phenyl, polyaniline, polyaramids, polyamides, polyhydrazides, polyurethanes, polyimides, polyazoles, polyazole ether ketone, polyureas, polyazines;

liquid-crystalline polymers, especially Vectra; and

inorganic polymers, examples being polysilanes, polycarbosilanes, polysiloxanes, polysilica, polysilicates, silicones, polyphosphazenes and polythiazyl.

In order to improve the performance properties further it is possible for fillers as well to be added to the membrane, especially proton-conducting fillers, and also additional acids. The addition can be made for example in step A), step B), step C) and/or step D). These additives, if they are in liquid form, can also be added after the polymerization according to step D).

Nonlimiting examples of proton-conducting fillers are sulfates such as: CsHSO_4 , $\text{Fe}(\text{SO}_4)_2$, $(\text{NH}_4)_3\text{H}(\text{SO}_4)_2$, LiHSO_4 , NaHSO_4 , KHSO_4 , RbSO_4 , $\text{LiN}_2\text{H}_5\text{SO}_4$, NH_4HSO_4 ,

phosphates such as $\text{Zr}_3(\text{PO}_4)_4$, $\text{Zr}(\text{HPO}_4)_2$, $\text{HZr}_2(\text{PO}_4)_3$, $\text{UO}_2\text{PO}_4 \cdot 3\text{H}_2\text{O}$, $\text{H}_8\text{UO}_2\text{PO}_4$, $\text{Ce}(\text{HPO}_4)_2$, $\text{Ti}(\text{HPO}_4)_2$, KH_2PO_4 , NaH_2PO_4 , LiH_2PO_4 , $\text{NH}_4\text{H}_2\text{PO}_4$, CsH_2PO_4 , CaHPO_4 , MgHPO_4 , HSbP_2O_8 , $\text{HSb}_3\text{P}_2\text{O}_{14}$, $\text{H}_3\text{Sb}_3\text{P}_2\text{O}_{20}$, polyacid such as $\text{H}_3\text{PW}_{12}\text{O}_{40} \cdot n\text{H}_2\text{O}$ ($n=21-29$), $\text{H}_3\text{SiW}_{12}\text{O}_{40} \cdot n\text{H}_2\text{O}$ ($n=21-29$), H_xWO_3 , HSbWO_6 , $\text{H}_3\text{PMo}_{12}\text{O}_{40}$, $\text{H}_2\text{Sb}_2\text{O}_{11}$, HTaWO_6 , HfNbO_5 , HTiNbO_5 , HTiTaO_5 , HSbTeO_6 , $\text{H}_5\text{Ti}_4\text{O}_9$, HSbO_3 , H_2MoO_4 ,

selenides and arsenides such as $(\text{NH}_4)_3\text{H}(\text{SeO}_4)_2$, UO_2AsO_4 , $(\text{NH}_4)_3\text{H}(\text{SeO}_4)_2$, KH_2AsO_4 , $\text{Cs}_3\text{H}(\text{SeO}_4)_2$, $\text{Rb}_3\text{H}(\text{SeO}_4)_2$,

oxides such as Al_2O_3 , Sb_2O_3 , ThO_2 , SnO_2 , ZrO_2 , MoO_3 , silicates such as zeolites, zeolites(NH_4^+), sheet silicates, framework silicates, H-natrolite, H-mordenite, NH_4 -anal-cine, NH_4 -sodalite, NH_4 -gallate, H-montmorillonite acids such as HClO_4 , SbF_5

fillers such as carbides, especially SiC , Si_3N_4 , fibers, especially glass fibers, glass powders and/or polymer fibers, preferably based on polyazoles.

These additives may be present in usual amounts in the proton-conducting polymer membrane, although the positive properties of the membrane, such as high conductivity, long life and high mechanical stability, ought not to be too greatly affected by adding excessive amounts of additives. In general the membrane after the polymerization according to step D) contains not more than 80% by weight, preferably not more than 50% by weight and more preferably not more than 20% by weight of additives.

Said membrane may also further comprise perfluorinated sulfonic acid additives (preferably 0.1-20% by weight, more preferably 0.2-15% by weight, very preferably 0.2-10% by weight). These additives lead to performance improvement, to an increase in oxygen solubility and oxygen diffusion in the vicinity of the cathode, and to a reduction in the adsorption of phosphoric acid and phosphate to platinum. (Electrolyte additives for phosphoric acid fuel cells. Gang, Xiao; Iijuler, I. A.; Olsen, C.; Berg, R. W.; Bjerrum, N. J. Chem. Dep. A, Tech. Univ. Denmark, Lyngby, Den. J. Electrochem. Soc. (1993), 140(4), 896-902 and Perfluoro-

sulfonimide as an additive in phosphoric acid fuel cell. Razaq, M.; Razaq, A.; Yeager, E.; DesMarteau, Darryl D.; Singh, S. Case Cent. Electrochem. Sci., Case West. Reserve Univ., Cleveland, Ohio, USA. J. Electrochem. Soc. (1989), 136(2), 385-90.)

Nonlimiting examples of persulfonated additives are:

trifluoromethanesulfonic acid, potassium trifluoromethanesulfonate, sodium trifluoromethanesulfonate, lithium trifluoromethanesulfonate, ammonium trifluoromethanesulfonate, potassium perfluorohexanesulfonate, sodium perfluorohexanesulfonate, lithium perfluorohexanesulfonate, ammonium perfluorohexanesulfonate, perfluorohexanesulfonic acid, potassium nonafluorobutanesulfonate, sodium nonafluorobutanesulfonate, lithium nonafluorobutanesulfonate, ammonium nonafluorobutanesulfonate, cesium nonafluorobutanesulfonate, triethylammonium perfluorohexanesulfonate, perfluorosulfonimides and Nafion.

The sheetlike structure is formed according to step B) by means of measures which are known per se (casting, spraying, knife coating, extrusion), being known from the prior art relating to polymer film production. Accordingly the mixture is suitable for forming a sheetlike structure. The mixture can accordingly constitute a solution or a suspension, the fraction of the components of low solubility being limited to quantities which allow sheetlike structures to be formed. Suitable supports are all supports which can be termed inert under the conditions. These supports include, in particular, films of polyethylene terephthalate (PET), polytetrafluoroethylene (PTFE), polyhexafluoropropylene, copolymers of PTFE with hexafluoropropylene, polyimides, polyphenylene sulfides (PPS) and polypropylene (PP).

To adjust the viscosity it is possible where appropriate to add water and/or a readily evaporable organic solvent to the mixture. This allows the viscosity to be adjusted to the desired value and makes it possible to facilitate the formation of the membrane. The thickness of the sheetlike structure is generally between 15 and 2000 μm , preferably between 30 and 1500 μm , in particular between 50 and 1200 μm , but this should not be taken to constitute a restriction.

The polymerization of the vinyl-containing sulfonic acid and, where present, vinyl-containing phosphonic acid in step D) takes place preferably by free-radical means. The free radicals may be formed thermally, photochemically, chemically and/or electrochemically.

By way of example, an initiator solution which comprises at least one substance capable of forming free radicals can be added to the mixture after the heating of the solution and/or dispersion according to step B). It is also possible to apply an initiator solution to the sheetlike structure obtained after step C). This can be done by means of measures known per se (e.g., spraying, dipping, etc.), being known from the prior art.

Suitable free-radical initiators include azo compounds, peroxy compounds, persulfate compounds or azoamides. Nonlimiting examples are dibenzoyl peroxide, dicumene peroxide, cumene hydroperoxide, diisopropyl peroxydicarbonate, bis(4-*t*-butylcyclohexyl)peroxydicarbonate, dipotassium persulfate, ammonium peroxydisulfate, 2,2'-azobis(2-methylpropionitrile) (AIBN), 2,2'-azobis(isobutyramidine) hydrochloride, benzopinacol, dibenzyl derivatives, methyl ethylene ketone peroxide, 1,1-azobiscyclohexanecarbonitrile, methyl ethyl ketone peroxide, acetylacetone peroxide, dilauryl peroxide, didecanoyl peroxide, *tert*-butyl per-2-ethylhexanoate, ketone peroxide, methyl isobutyl ketone peroxide, cyclohexanone peroxide, dibenzoyl peroxide, *tert*-butyl peroxybenzoate, *tert*-butyl peroxyisopropyl carbonate,

FULLERENE-BASED ELECTROLYTE FOR FUEL CELLS

CROSS-REFERENCES TO RELATED APPLICATIONS

[0001] This patent application is a continuation-in-part of U.S. patent application Ser. No. 10/867,380 filed Jun. 12, 2004, which claims the benefit of U.S. Provisional Application No. 60/477,971, filed Jun. 12, 2003, and U.S. Provisional Application No. 60/500,603, filed Sep. 5, 2003.

STATEMENT REGARDING FEDERALLY-SPONSORED RESEARCH AND DEVELOPMENT

[0002] This invention was made with Government support under Government Contract No. DAAD19-03-C-0024, awarded by the United States Department of Defense. The Government has certain rights in the invention.

BACKGROUND OF THE INVENTION

[0003] The present invention relates to polymer electrolyte membranes for use in fuel cells, and more particularly, to the utilization of fullerene materials for enhancing the low relative humidity proton conductivity properties of such polymeric membranes.

[0004] A steadily increasing demand for portable electric power has stimulated interest in the development of more efficient and more powerful fuel cell devices. A polymer electrolyte membrane (PEM) fuel cell is a strong candidate as a portable power source for commercial applications primarily because of its low weight and high power density.

[0005] The operation of a PEM fuel cell relies upon the proton conductivity properties of a polymeric membrane positioned between the two electrodes of the cell, to transport protons internally from one electrode to the other. The membrane must also have no electronic conductivity, good chemical and mechanical stability, and sufficient gas impermeability to prevent cross over of the fuel. For many years now, the membrane of choice has been a sulfonated perfluoro polymer known as Nafion®, commercially available from DuPont.

[0006] The major drawback to Nafion® as the ideal polymer electrolyte membrane in fuel cells is that its proton conductivity depends on the water content in the membrane, in which proton transport is based on the diffusion of hydronium ion (H_3O^+). In order to retain its high proton conductivity, Nafion® membrane requires the use of pre-humidified gases at an operating temperature under 80° C. Such requirements considerably increase the cost, size and complexity of PEM fuel cells using Nafion®. Nafion® membranes cannot perform under dry or low relative humidity conditions nor above the boiling point of water, despite the faster chemical reaction and increased output that would result from the higher temperature. Furthermore, operating at the lower temperature required by Nafion® increases the risk of carbon monoxide poisoning of the fuel cell catalyst.

[0007] Various attempts have been made to develop water-free proton conductive membranes for PEM fuel cells that do not have the low temperature and high relative humidity requirements of Nafion®. One such attempt, for example, is described in the Hinokuma et al. U.S. Pat. No. 6,495,290,

issued Dec. 17, 2002, incorporated herein by reference. The proton conductors employed by Hinokuma et al. are based on fullerene derivatives containing acidic functional groups such as $-OH$ or $-SO_3H$, and are designed to operate under dry conditions over a wide range of temperatures. The proton conductors are described as being either compacted powder of the fullerene derivatives, or mixed with a small amount, generally 20 weight percent or less, of a film-forming polymeric material, such as polytetrafluoroethylene, polyvinylidene fluoride or polyvinyl alcohol. The patent cautions against employing the polymer in amounts any greater than 20 weight percent, at the risk of degrading the proton conductivity of the fullerene derivative. Furthermore, there is no hint in the Hinokuma et al. patent of using the fullerene derivative in combination with Nafion®.

SUMMARY OF THE INVENTION

[0008] The present invention resides in the discovery that proton conductive fullerene materials, as well as non-conductive fullerene materials that nevertheless facilitate the overall proton conductivity of a conductive membrane, including but not limited to the fullerene derivatives described in Hinokuma et al., U.S. Pat. No. 6,495,290, can be used in minor amounts, even as low as about 1% by weight, to enhance the low relative humidity proton conductivity properties of a variety of polymeric materials, even including Nafion® itself. The proton conductive fullerene materials used in the present invention contain either bound water, or a plurality of functional groups with lone pair electrons, or a combination thereof, and may be incorporated into the polymeric material by doping or by mechanical mixing or by chemical reaction forming covalent bonds. The resulting proton conductors may be used as polymer electrolyte membranes in fuel cells operative over a wide range of relative humidity conditions and over a wide range of temperatures from below room temperature to above the boiling point of water.

[0009] The present invention includes the use of proton conductive and facilitating yet non-conductive fullerene materials to enhance the low relative humidity proton conductivity properties of polymeric materials. It also includes the proton conductors resulting from such use, as well as fuel cells employing such proton conductors.

BRIEF DESCRIPTION OF THE SEVERAL VIEWS OF THE DRAWINGS

[0010] FIG. 1 is a graph showing the results of measuring the proton conductivities as a function of relative humidity at 30° C. of a bare Nafion® membrane (plot a) in comparison with Nafion® membrane doped with 1% by weight of various fullerene materials (plots b, c and d) and polycethylene oxide-fullerene materials composites (plots e and f).

[0011] FIG. 2 is a graph showing the polarization curves of a PEM fuel cell measured at 120° C. under 25% relative humidity with a bare Nafion® membrane (plot a) in comparison with Nafion® membrane doped with 1% by weight of fullerene materials (plots b and c); and

[0012] FIG. 3 is a sectional view showing a fuel cell that employs a proton conductor in accordance with the present invention.

brane/catalyst/electrode interface. The relative amounts of these monomers allow controlled swellability, and allow the product to be rigid, or solid and elastic.

[0020] Cross-linked materials may be formed by using γ -irradiation or thermal irradiation. For example, ionising radiation, e.g. 90 MeV gamma radiation from a cobalt 60 source may be used, in which case no cross-linking agent needs to be added. Nevertheless, it is possible to control the properties of the final material by the addition of:

[0021] (c) a chemical cross-linking agent such as allyl methacrylate or ethylene glycol dimethacrylate, and

[0022] (d) a chemical initiator such as AIBN (azoisobutyronitrile) or azobiscyclohexanecarbonitrile.

[0023] If the material is to be thermally initiated and cross-linked, then components (c) and (d) above become essential to the process.

[0024] Such hydrophilic materials may be converted to electrically active systems, cationic or anionic, by the addition of:

[0025] (e) an electrically active molecule held within the matrix of the hydrophilic polymer; or

[0026] (f) an electrically active comonomer.

[0027] The electrically active component can be based either upon an acid, e.g. a sulphonic acid (SO_3H), phosphoric or phosphonic acid, or an alkali, e.g. a compound providing OH^- ions such as KOH, NaOH or ammonium hydroxide or vinylbenzyltrimethylammonium chloride. The preferred monomer is 2-acrylamido-2-methyl-1-propanesulphonic acid (AMPS), styrenesulphonic acid (SA), vinylsulphonic acid (VA) or SOMA. It is also possible that component (a) or (b) may be modified so that it also functions as component (f).

[0028] An electrically active molecule (e) may be held within the matrix by steric interference as an alternative to, or in addition to, chemical bonding. Addition of a swelling liquid (e.g. ethyl alcohol) to the hydrophilic polymer can cause greater swelling than with water. Ionically active molecules dissolved in the swelling liquid will exchange for water by diffusion and the polymer will shrink, thereby entrapping the molecules within the matrix. Such an effect is observed with 50:50 MMVA-VP copolymer and ionic molecules dissolved in ethyl alcohol.

[0029] One or more types of ionically active molecules can be introduced into the matrix using this method. Subsequent activation of the material by gamma-irradiation may cause a reaction between introduced molecules, to form a molecule larger than those entrapped by steric interference, and/or a binding reaction of an introduced molecule with the polymer matrix.

[0030] In a solid polymer electrolyte as used in any form of PEM system, the ionic conduction (C_i) should be very much greater than the electronic conduction (C_e). A C_i/C_e ratio of less than 0.1 is desirable for successful operation.

[0031] Polymers suitable for use in the invention may be produced by polymerisation of the monomers and water or another liquid in which component (f) is soluble and with which the other components are miscible. The involvement of water is not fully understood, but as an acidic or alkaline

solution it apparently acts as a comonomer and mediates the introduction of the acid or alkali moieties into the cross-linked polymer structure. After polymerisation, some or all of the water may be removed, but rehydration does not necessarily give the product obtained before drying.

[0032] Considerations that should be given to the materials include their hydrophilicity, for control of water and gas permeability independent of the electrical properties, and their cross-linking, for stability; the use of sulphonic acid, phosphoric acid, etc; the use of alkaline side chain for alkaline fuel cells; and the use of water or alcohol to carry the electrically active moiety into the polymer, the polar solution acting (unexpectedly) as a co-monomer. As the polymer, AN-VP plus AMPS is preferred, but other suitable monomer combinations include MMA-VP and MMA-HEMA.

[0033] In general, the catalyst may be prepared in several ways:

[0034] (a) as a slurry of solid particles in a support fluid, as an 'ink' which is then applied uniformly or as a predetermined pattern as a spray, or as a liquid by painting or spreading by any suitable technique including a doctor blade or 'ink jet' process;

[0035] (b) as a dry powder, such that the particles can be distributed uniformly or in a predetermined pattern to a surface by any suitable means including electrostatic spraying or rubbing into a textured surface; or

[0036] (c) as a solution or reactant which is capable of being delivered as a liquid and precipitated or otherwise chemically deposited on the surface.

[0037] Preferred methods of the invention include the following Processes 1 to 7. ITM-SPE is given for the purpose of illustration only, and other materials may be used instead.

Process 1

[0038] A preformed catalyst layer is contacted, e.g. under pressure, with the surface of fully hydrated cross-linked hydrophilic ionically active material (the ITM-SPF). These materials are characterised by being soft and elastic when hydrated and in this form the surface is sufficiently soft and deforming to allow the catalyst particles to be brought into intimate contact by pressure alone. Thus two electrodes defining the boundaries of the cell are coated with catalyst by any suitable method, the catalyst is dried or cured to adhere to the electrode materials and the electrodes are pressed against the surface of a ITM-SPF membrane. Alternatively the coated electrodes are held apart by a fixed distance and the ITM-SPE material, initially dehydrated or pre-stressed to reduce its thickness, is introduced into the gap. On hydration, the ITM-SPE swells and makes contact with the electrodes, so imparting the necessary interfacial pressure to maintain good contact.

[0039] Alternatively, the catalyst layer can be applied to the hydrophilic surface and held in place by electrode surfaces pressed against the catalyst.

Process 2

[0040] In this procedure, solid catalyst particles are included in the surface of the ITM-SPE during polymerisation by a transfer process, e.g. catalyst is deposited onto a

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REMARKS/ARGUMENTS

Claims 29-35, 43-46, 48-49 and 51 are rejected under 35 U.S.C. 102(e) as being anticipated by Kim et al. (Pub. No.: US 2003/0099874).

The Examiner states that "As to claim 29, Kim et al. shows a method for manufacturing an electrical circuit comprising a step of forming at least a part of the electrical circuit by impregnating a conductive polymer [0039] (Fig. 1 and related text) exhibiting p-type [0043-0046] conduction or n-type conduction [0043-0046] (conductive particles with Boron and Phosphorous exhibit p-type and n-type conduction in [0043]) in a receptive layer [0047] (porous support or micro channels is used a receptive layer)".

Specifically, the Examiner states that "conductive particles with Boron and Phosphorous exhibit p-type and n-type conduction in [0043]". In the response to arguments to contrary, the Examiner takes the position that the Kim conductive polymer "inherently" exhibits p-type or n-type conduction.

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Applicants submit that "inherent" anticipation requires certainty - not just possible or probable. (See *In re Oelrich and Divigard*, 212 USPQ 323, 326 (CCPA 1981).

However, paragraph [0042] of Kim et al. describes as follows: "Here, as the ionic conductive material, at least one selected from the group consisting of a heteropoly acid of formula (1) below, a phosphoric acid of formula (2) below, sulfuric acid and salts of these materials are used". Accordingly, it is clear that the compound represented by formula (1) or formula (2) is an ionic conductive material.

Kim et al. employs $\text{H}_3\text{PW}_{12}\text{O}_{40} \cdot \text{nH}_2\text{O}$ (containing phosphorus P) as a heteropoly acid in EXAMPLES of Kim et al. ([0056]-[0073]). Enclosed above is a copy of Column 21 of Kiefer et al USP 7,332,530. It is disclosed in US7332530 that $\text{H}_3\text{PW}_{12}\text{O}_{40} \cdot \text{nH}_2\text{O}$ is a proton-conduction material, namely, an ionic conductor (Col.21, line 33).

An ionic conductive material is a material in which the charge carrier is "ions" and conduction due to electrons or positive holes is extremely limited.

In the polymer electrolyte of Kim et al., above (ion conducting) inorganic conductive particles are impregnated into microchannels formed by aggregation of polar portions

of the ionic conductive polymer membrane, and/or between backbones of the ionic conductive polymer membrane (ABSTRACT).

Since the conductive polymer membrane of Kim et al. is used as an electrolyte of a fuel cell, the conductive polymer is also an ionic conductive material as taught, for example, in the ABSTRACT.

It is well known that n-type conduction or p-type conduction must be suppressed very low in an electrolyte of a fuel cell in order to avoid cancellation of the voltage generated between the both surfaces of the electrolyte due to leakage of the electric charge by the n-type conduction or the p-type conduction. Namely, the electrolyte of a fuel cell must be an ionic conductive material instead of an n-type conductor or a p-type conductor.

Since almost pure ionic conduction of the electrolyte is a common feature for every kind of battery, such as a fuel cell, it is not usually mentioned specifically in technical documents, for example patent documents, however, there are some documents in which the ionic conductive nature of an electrolyte is clearly mentioned.

For example, Ecer US20080003481 (see copy of page 5 above) discloses a solid electrolyte for a fuel cell and

teaches as follows: "Here the solid electrolyte maybe an oxide, sulfide, bromide, chloride or any other substance that allows charged particle conduction, but little or no electronic conduction ([0073], under lines were added by the Applicants)."

US20060024546 (copy of [0005]-[0006] enclosed above). discloses a fuel cell employing the same polymer electrolyte Nafion® as employed by Kim et al. ([0005]-[0006]) and teaches as follows: "The operation of a PEM fuel cell (PEM stands for polymer electrolyte membrane) relies upon the proton conductivity properties of a polymeric membrane positioned between the two electrodes of the cell, to transport protons internally from one electrode to the other. The membrane must also have no electronic conductivity, good chemical and mechanical stability, and sufficient gas impermeability to prevent cross over of the fuel. For many years now, the membrane of choice has been a sulfonated perfluoro polymer known as Nafion®, commercially available from DuPont ([0005], under lines were added by the Applicants). It further teaches: "The selection will generally be made based upon the other requisite properties for a fuel cell polymer electrolyte membrane, such as no electronic conductivity, good chemical

and mechanical stability, and sufficient gas impermeability to prevent cross over of the fuel ([0018], under lines were added by the Applicants)."

Although, only n-type conduction (electronic conduction) is mentioned above, also p-type conduction (hole conduction) must not be contained in the conduction of the electrolyte of a fuel cell.

N-type conduction or p-type conduction, if any in a polymer electrolyte of a fuel cell, should be very small as taught in WO2005124915 (corresponding Highgate et al US 2008/0063920 provided above at [0030]: "In a solid polymer electrolyte as used in any form of PEM system, the ionic conduction (C') should be very much greater than the electronic conduction- (C_e). A C_e/C' ratio of less than 0.1 is desirable for successful operation."

Therefore, not only is Kim et al different from the present invention as now claimed, it would not be obvious to modify Kim or use the teachings. One of ordinary skill in the art would not be motivated to use the polymer electrolyte of Kim et al. to fabricate the electrical circuit of the present Application by impregnating a conductive polymer which must be a p-type conductor or an n-type conductor in a receptive layer as requested in

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(amended) claim 29 of the present Application. As detailed above, the problems are different and the nature of the solution to problems is not analogous.

Further, the technical field of Kim et al. is quite different from that of the present Application. The U.S. Cl. of Kim et al. is 429/33 and 521/27 where:

429: Chemistry: Electrical current producing
apparatus, product and process; and

521: Synthetic resins or natural rubbers,

while the U.S. Cl. of the present Application is
257/40 where:

257: Active solid-state devices.

Accordingly, one of ordinary skill in the art would not refer the conductive polymer of Kim et al. to fabricate the electrical circuit of the present Application. Even combining Kim with the other art relied on for certain features, does not bridge the missing teaching.

Therefore, the method for manufacturing an electrical circuit of the present Application is not obvious over Kim et al, alone or in combination with the other art.

Accordingly, Claim 29 as now amended is not shown or suggested by the art, alone or in combination. Withdrawal

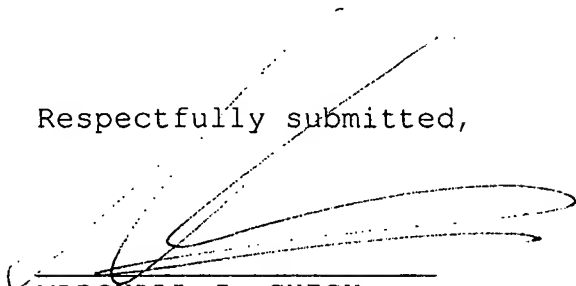
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of the rejection along with that of Claims 30 - 51,
dependent on Claim 29, is therefore requested.

In view of the above, the rejections are avoided.
Allowance of the application is therefore respectfully
requested.

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Respectfully submitted,



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